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Comment on acp-2021-96

Anonymous Referee #1

Referee comment on "The impact of aerosol size-dependent hygroscopicity and mixing state on the cloud condensation nuclei potential over the north-east Atlantic" by Wei Xu et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-96-RC1, 2021

This study presents data collected from the Mace Head field operational site and presents an analysis of the influence of the size-dependent hygroscopicity and mixing state of aerosols for the prediction of the CCN number. The authors compare CCN number closure using 6 different methods on aerosol concentrations measured over Northeast Atlantic. The work provides significant long-range measurement of CCN from one site. The sampled air masses are divided into sectors, including polluted continent, clean oceanic, and mixtures for both high and low biological level of activity. It was concluded that for low SS the mixing state plays an important role while for high SS the hygroscopicity is size dependent.

The paper is appropriate for submission to ACP. The issue of N_{CCN} closure is of scientific importance and thus the work contributes new data and methods of scientific significance to the field. There are several grammatical errors and typo through the manuscript. There are some questions regarding the approach and interpretation of the analytical methods D-F. There are also additional questions regarding the relevance/reliance on data provided in the supplemental document for the understanding of the main manuscript.

Overall, the manuscript and contents continue the important scientific discussion about N_{CCN} prediction and should be published. This reviewer congratulates the authors on synthesizing a significantly large and complex CCN measurement and prediction data set. There are several unique features that can be discerned over a long period of time and the authors have overall presented a cohesive narrative that expands our knowledge of CCN prediction and the aerosol-indirect effect.

Below are some major and minor comments that are suggested to improve the clarity of the work.

Major Comments:

- A major conclusion (L531) is that the mixing state varies at different SS. However this is mainly true for the Clean and Polluted air masses, as is focused on in the paper. As suggested, these air masses have uniform properties (Table 1). Is the same conclusion true of the mixed air masses? Or is it possible that mixing state impacts the N_{CCN}at both high and low SS in the mixed air datasets? If so, the main conclusion should emphasize to the reader that mixing state has minimal effects on $N_{CCN}$when the air masses can be separated.
- There are concerns with the logic of method C for analysis. Given the authors state that the Aitken mode is that which is most sensitive to changes in hygroscopicity, why choose a size (165 nm) in the accumulation mode? This proliferates to the efficacy of methods for inferring mixing states (Fig 6, middle panel). If method C, is based on a smaller particle diameter (one in the Aitken mode) will it not become more efficient?
- Page 16 line 392: The authors claim that there was no statistically significant difference between predictions obtained using method D and method E. However, an analysis for that is not shown. Ideally hypothesis testing by performing p-value estimation or student t-test, the statistical significance of the results can be determined. Statistical significance can probably not be determined simply by comparing the RRSE or R-scores against each other obtained using the 2 methods.
- Page 22 line 560: It does not seem that the effectiveness of method E with respect to methods D and F follow a definitive pattern. The relative error for the continental cases seem to have an almost 0 correlation with the ambient type (polluted, mixed, or clean). And the relative error for the marine cases have both positive and negative correlations with the ambient types. Therefore, on what basis can it be inferred that the effectiveness generally decreases with increasing SS? The explanation seems to be missing or unclear.
- The use of both the Pearson score and RRSE is unclear. How are the RRSE values interpreted and what do they represent for individual closures? Pearson's coefficient is self-sufficient for quantifying the uncertainties between measured and estimated/predicted values of a variable, then what purpose are RRSE scores serving?
- This reviewer has some reservations about the calibration of the instruments. How often was the CCNC calibrated? The frequency of calibration is also associated with the +/- 0.03% potential SS drift; the longer the times between calibration the greater potential for greater SS uncertainty. Furthermore, it may be of interest to include an explanation of how the HTDMA setup and RH were calibrated/validated as the derived hygroscopicity is applied to 3 of 6 methods. The conclusion can also benefit with a summary of the advantage/disadvantages of each method.
- Page 7 line 215: The criteria for considering the uncertainty of 20% in the hygroscopicity seems unclear. Specifically, how do the stated uncertainties in the other measurements relate to the assumed uncertainty of 20% in ? Is there a mathematical explanation behind this, or was this empirically chosen based on the findings of the authors or previous studies?
- The author, throughout the paper, makes many repeated references to figures and tables that are found in the supplemental section. Considering their importance, wouldn't it be more appropriate to incorporate them in the paper itself?
- Lastly, a summary lookup table of the effects of each method would be useful to have in the main paper. This can be appended onto Table 3 in a column that shows the relative error, or approximate magnitude of over/underestimation.

Minor Comments:

Line 15-16: Please check method description. “Method C utilized size dependent…” Isn’t this D?

Line 27: GF-PDf or GF-PDF?

Line 33: supersaturation vs super-saturation (line 11)

Line 51: nss-sulfate is not defined until line '154.

Line 147: arbitrary? Mixing State can be quantified. Please see Riemer, N., et al

Line 159: the letter d is missing from "and", “ammonium nitrate an sulfate”

Line 188: Please be aware that the font size and notation in the subscripts is not consistent. Similar inconsistencies can be found on other pages as well (pg 7, 8, so on and so forth).

Table 1: It is not clear in the criteria in the table if it is and/or. For example, BC< 15 ng AND WD from? Or is it one or the other?

Line 205: Please reword “were taking into”. Take into?

Page 8: The alignment of the columns in Table 2 is awkward. Perhaps centering?

Table 2: Is a reference for the density of organics required? What is the justification for 1400.

Line 239-240 why is the upper limit 500nm? Is 500nm the limitation in the DMA measurement?

Line 269: Change “In method F” to “In method E”.

Line 339 lend support “to”?

Line 343: Typo. Change to “Respectively”.

Line 343: HB and LB not defined.

Llne 355: "Fig 4&-5"?

Line 356: This range is peculiar. The minimum value of Pearson’ coefficient does not seem to be 0.85 (more like 0.76 in fig. 4 and 0.65 in fig. 5).

Line 361: Sentence is unclear.

Line 373, 380-381: Please double check ranges. The slope and R minimum and maximum values mentioned here do not match the ones written on the panels in fig. 4.

Line 423: The word “eith” is likely misspelt.

Line 454 “That indicated the potential over-estimation of 30% to 50% by using κ of 0.3 in highly polluted air masses” Is kappa or Nccn overestimated 30~50%?

Line 473: Change "compare" to compared

Line 485 “To conclude, using κ of 0.3 achieved reasonable closure in Polluted-H but resulted in significant over-estimation in Polluted-L by up to 50 to 60% and using bulk PM1 chemical composition **enabled to achieve** closure in Polluted-H, but showed over-estimation in Polluted-L.” What is enabled to achieve? Simply achieved?

Line 539: verb tense? Commonly adapted practice(s).

line 541: Please rephrase ‘pointing size mattered most at CCN activation.’ Indicating that?

Line 589: Reduced the error by 80% with respect to what?

REFERENCES:

Riemer, N., et al. "Aerosol mixing state: measurements, modeling, and impacts." *Reviews of Geophysics* 57.2 (2019): 187-249.